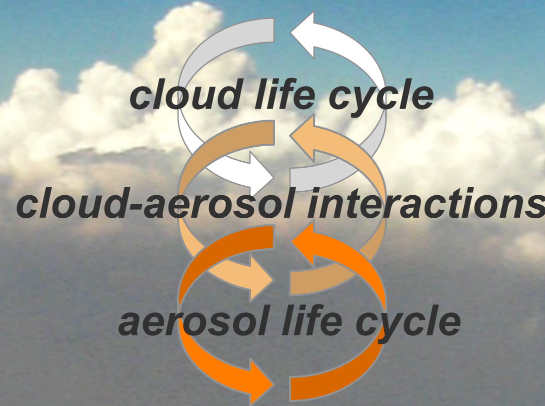

Modeling Secondary Organic Aerosols and Aerosol Radiative Forcing using the Volatility Basis Set Approach in WRF-Chem

Manish Shrivastava, Jerome Fast, James Barnard, and Rahul Zaveri

11th WRF User's Workshop, June 21-25, Boulder CO

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Motivation

Uncertainties in Organic Matter:

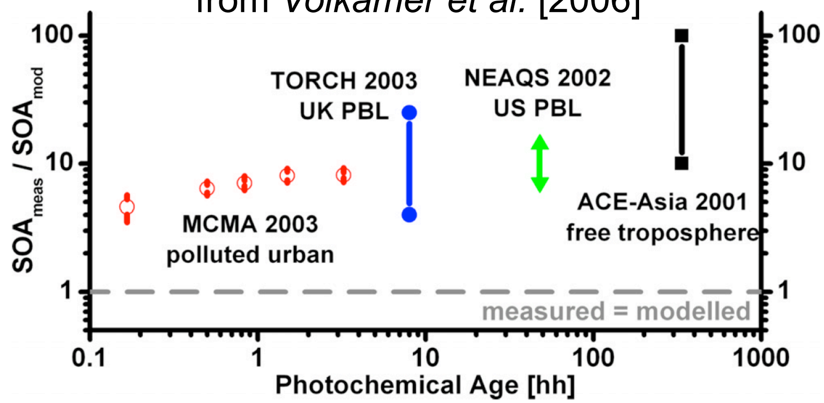
- Comprises ~50% of submicron mass worldwide [*Zhang et al.* 2007], and analyses of aerosol mass spectrometer data suggests it is comprised of mostly oxygenated material [*Jimenez et al.* 2010]
- Simulated organic matter mass usually too low because SOA formation is not adequately represented by models

WRF-Chem:

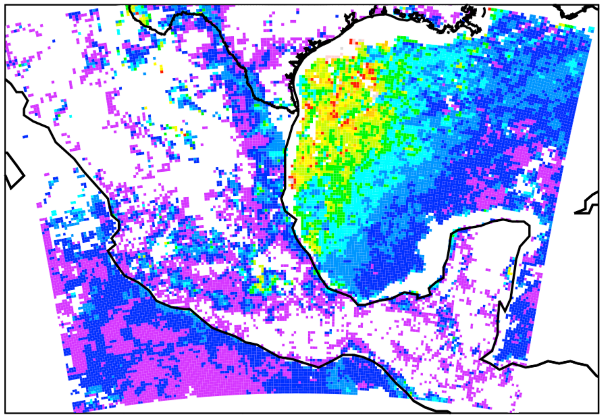
- SOA not treated in MOSAIC – assume organic matter is nonvolatile POA
- SOA treatment coupled with MOSAIC being tested and evaluated - likely available for next release of WRF
- Link new organic species to the aerosol optical property module (direct radiative forcing) & cloud-aerosol interaction modules (indirect radiative forcing)

Measured vs Modeled SOA

from *Volkamer et al.* [2006]



Average AOD during March 2006



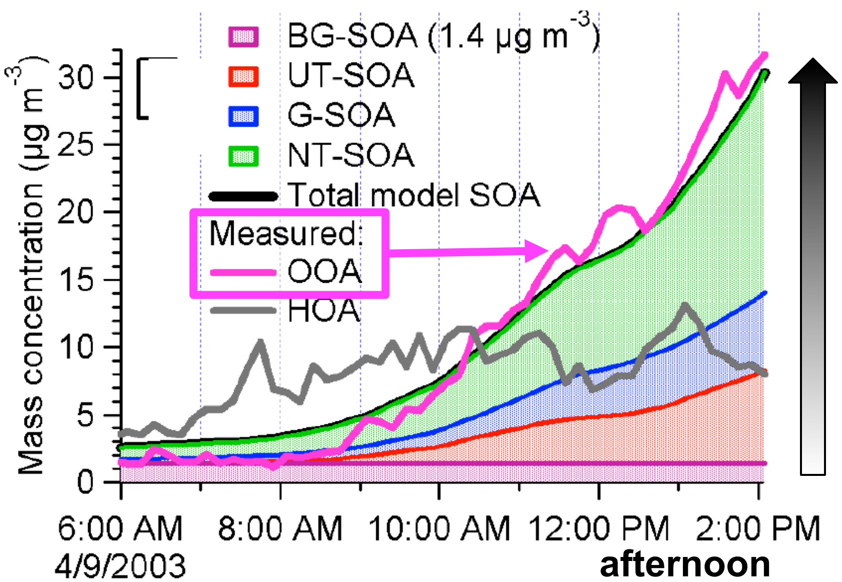
What is the impact of uncertainties in SOA on direct and indirect radiative forcing for climate?

Recent Modeling

- Simulated organic aerosol mass has improved recently: SOA_{model} ~ $SOA_{estimated}$
- But, many assumptions are employed by new treatments that cannot yet be tested

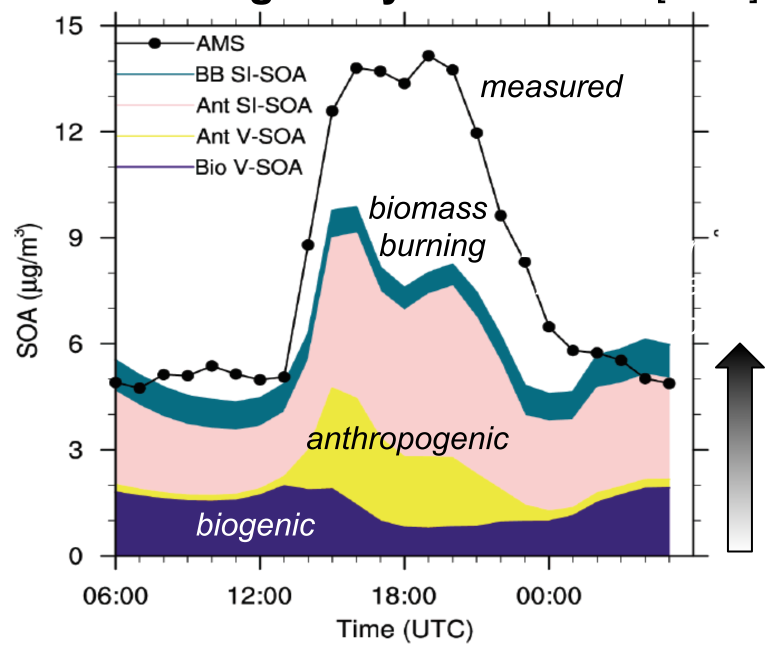
Examples

Box Modeling Study - *Dzepina et al.* [2009]



compared old and new approaches using AMS data in Mexico City

3-D Modeling Study - *Hodzic et al.* [2010]



used volatility basis set approach in Chimere model for Mexico City [*Donahue et al*. 2006; *Robinson et al.* 2007; *Shrivastava et al*. 2008]

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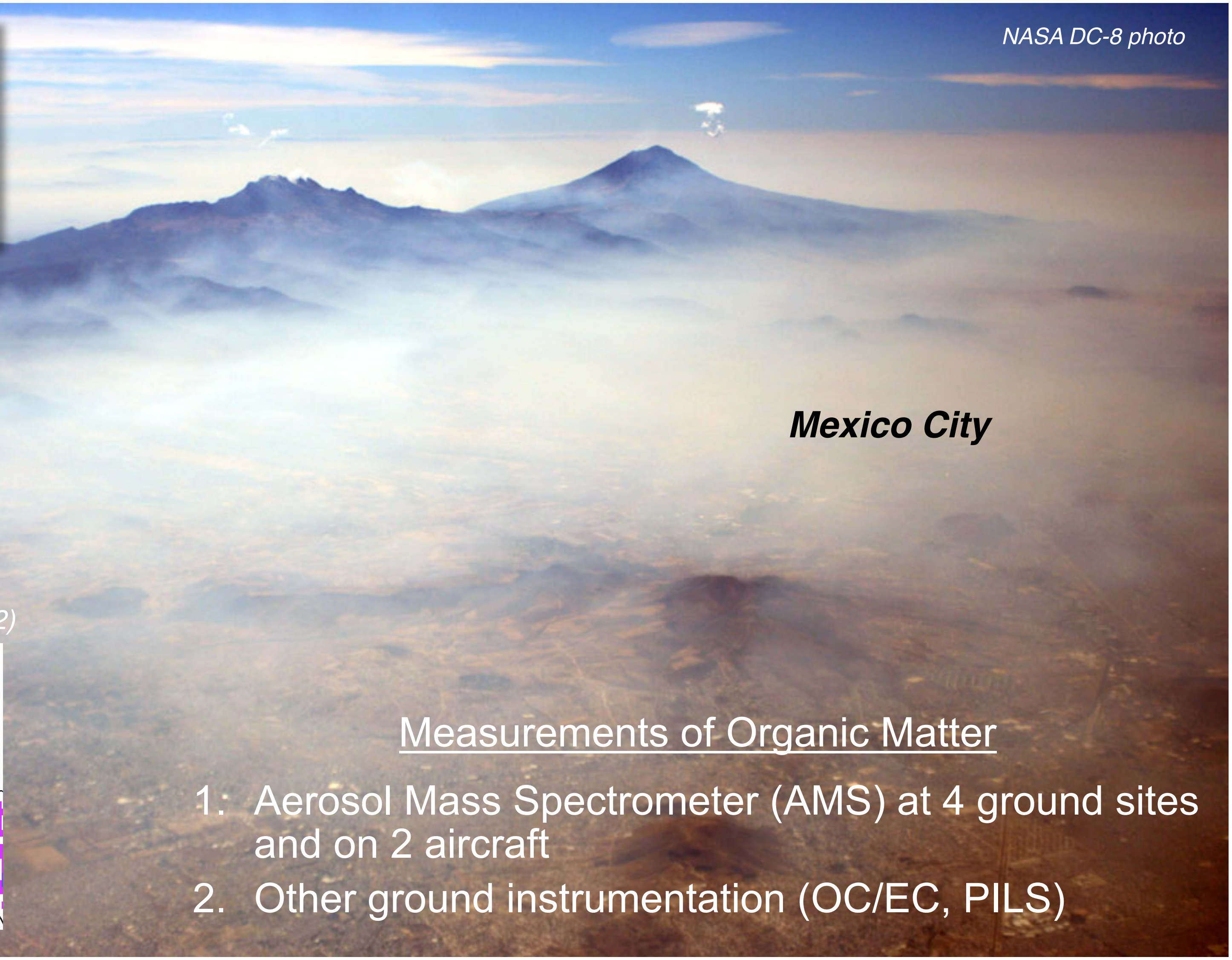
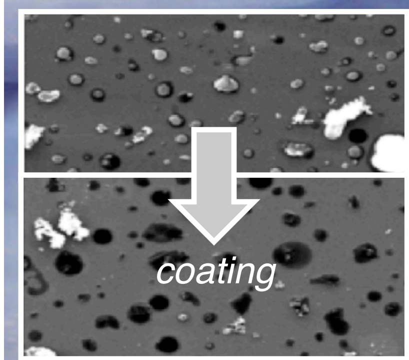
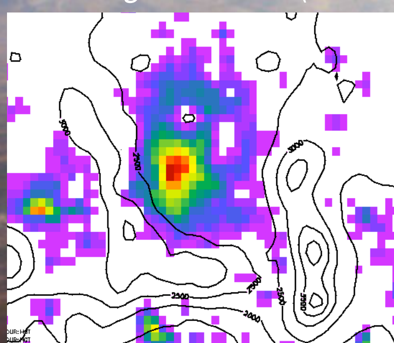
Testbed Case for SOA Treatment

Megacities Initiative: Local and Global Research Observations





2006 Emission Inventory From Miguel Zavala (MCE2)



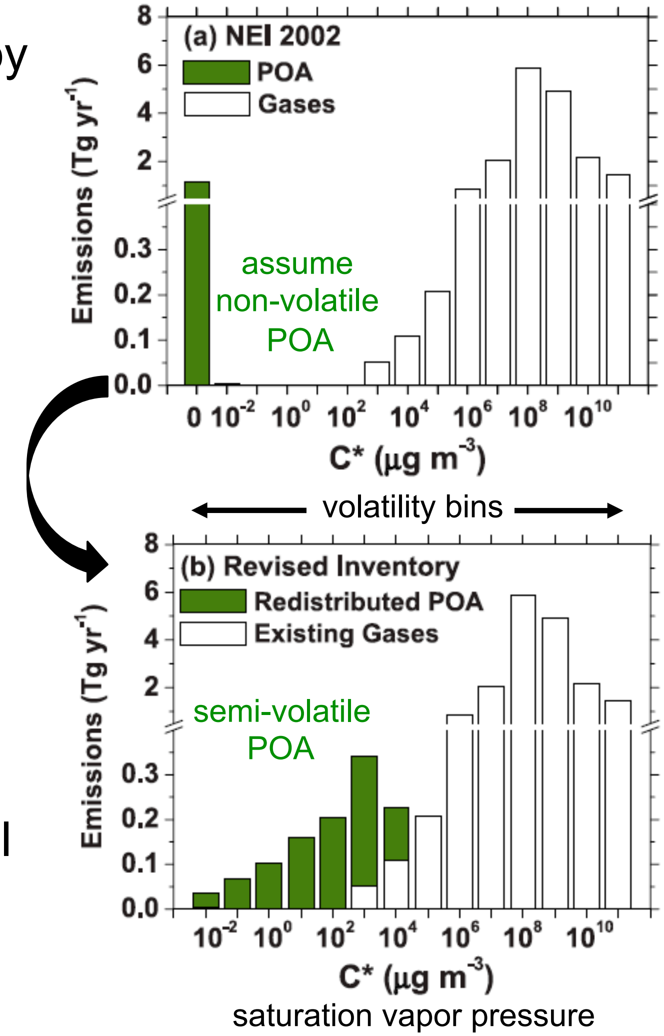
Measurements of Organic Matter

1. Aerosol Mass Spectrometer (AMS) at 4 ground sites and on 2 aircraft
2. Other ground instrumentation (OC/EC, PILS)

Our Approach

Organic Aerosol Treatment:

- Modified *Robinson et al*. [2007] volatility basis set by adding 2 oxygen atoms per generation of oxidation
- # of volatility bins: 9 for **fresh** and 8 for **aged**
- Separate volatility species for **fossil** and **biomass burning** sources
- Predict both **oxygen** and **carbon** mass for each volatility species to obtain O:C ratios
- Traditional anthropogenic and biogenic SOA (4-product VBS set) using yields from *Tsimpidi et al*. [2010] with no further aging
- prognostic SOA species: currently **380 for 4 size bins**, (684 for 8 size bins)
- Coupled with SAPRC-99 gas-phase mechanism (*Pablo Saide, U. Iowa*) and MOSAIC aerosol model
- Dry deposition for all species treated the same



Aerosol Optical Properties:

- For now, assume new organic species all have the same refractive index, density, etc.

Preliminary Results

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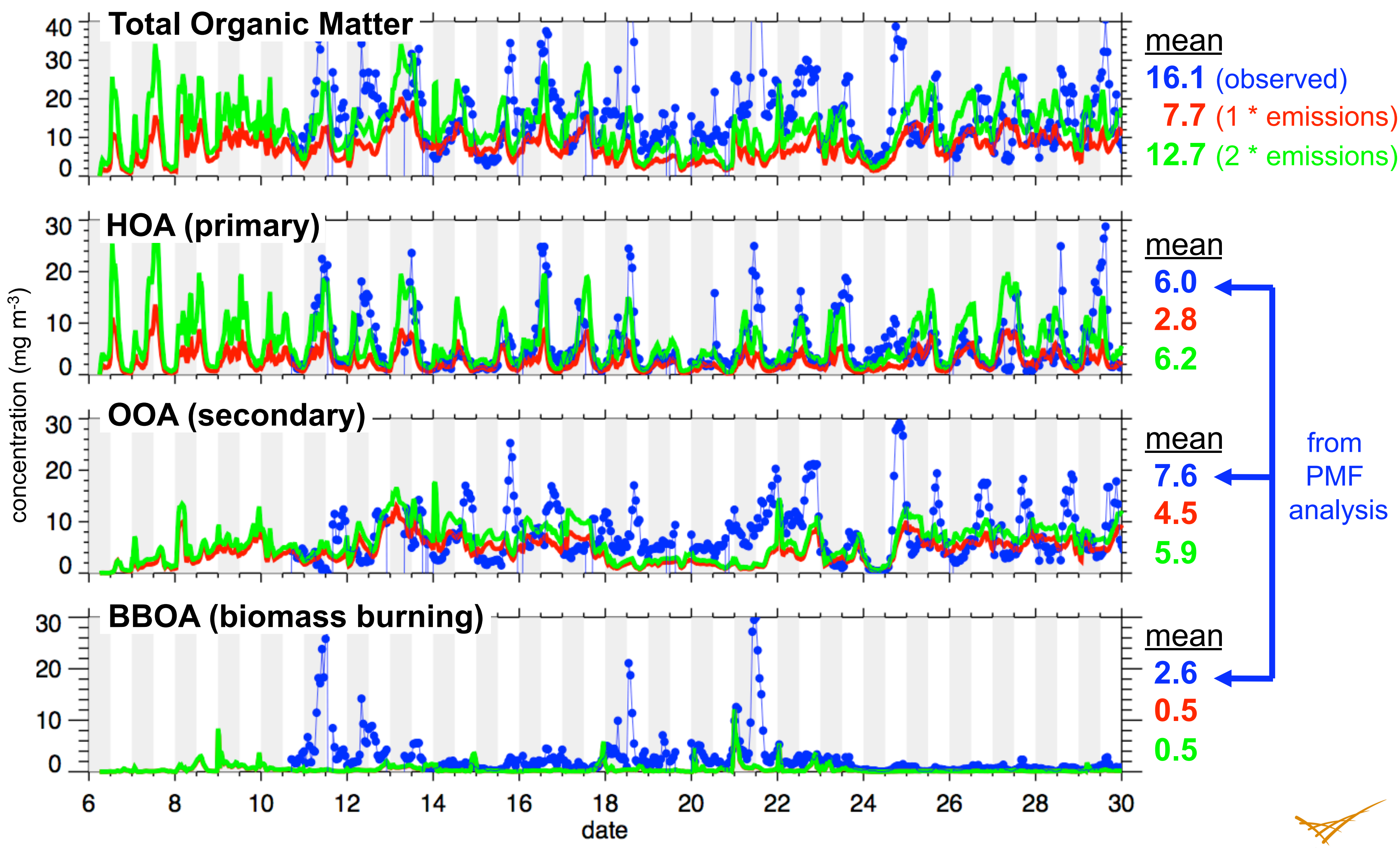
Temporal Variability in OM at T0 Site



Data and PMF analysis from Jose Jimenez and Allison Aiken (Univ. CO)

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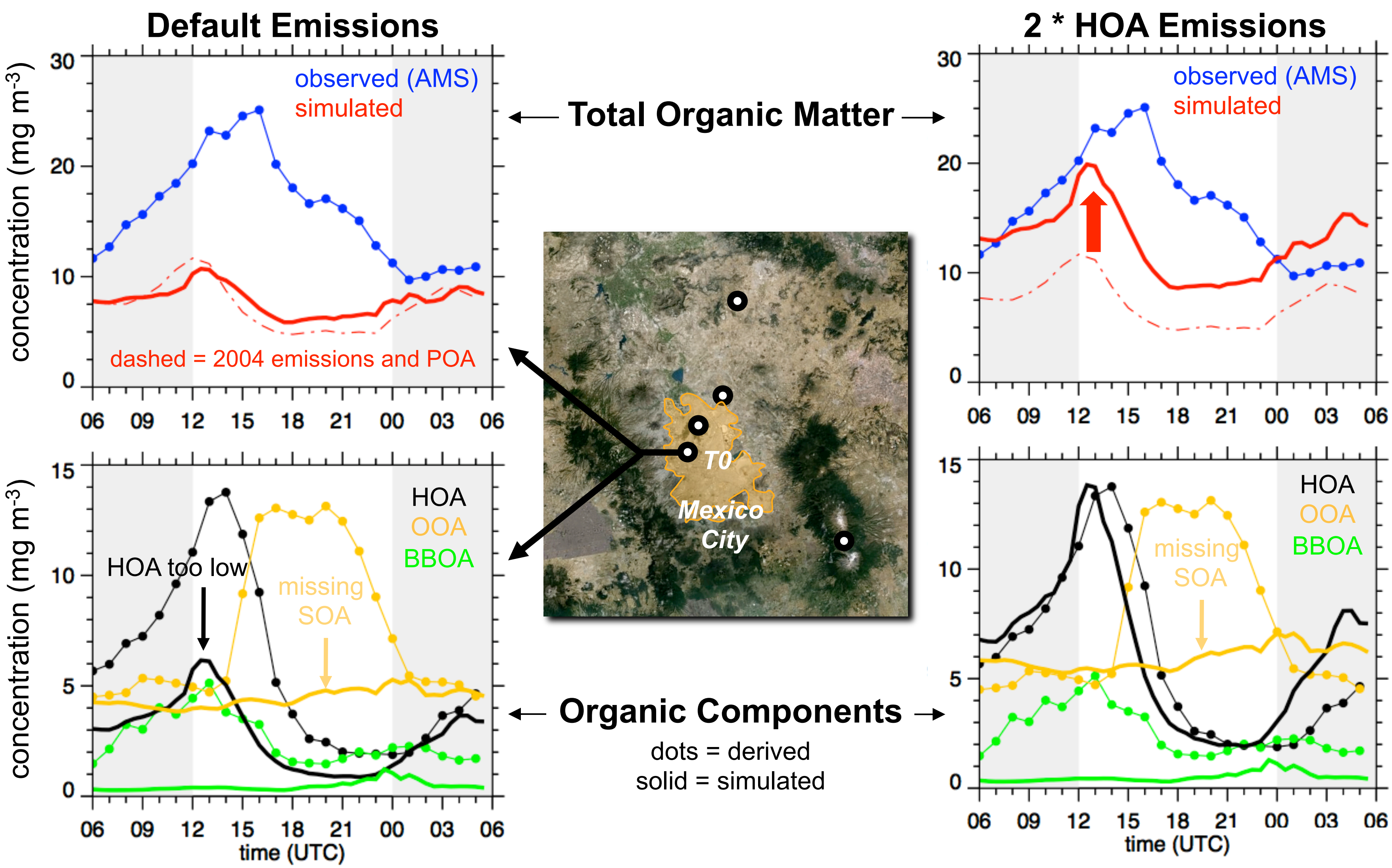
Diurnal Average OM: T0 Site



Data and PMF analysis from Jose Jimenez and Allison Aiken (Univ. CO)

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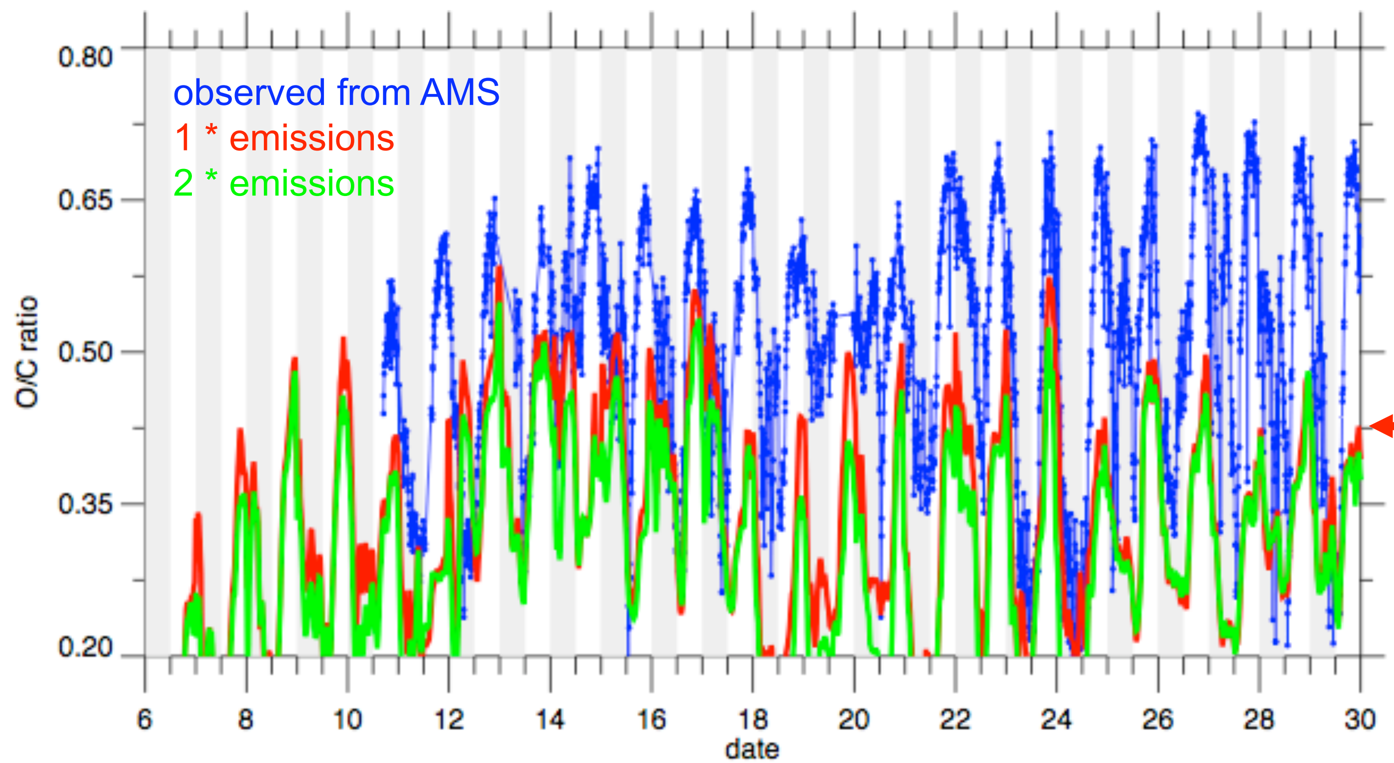
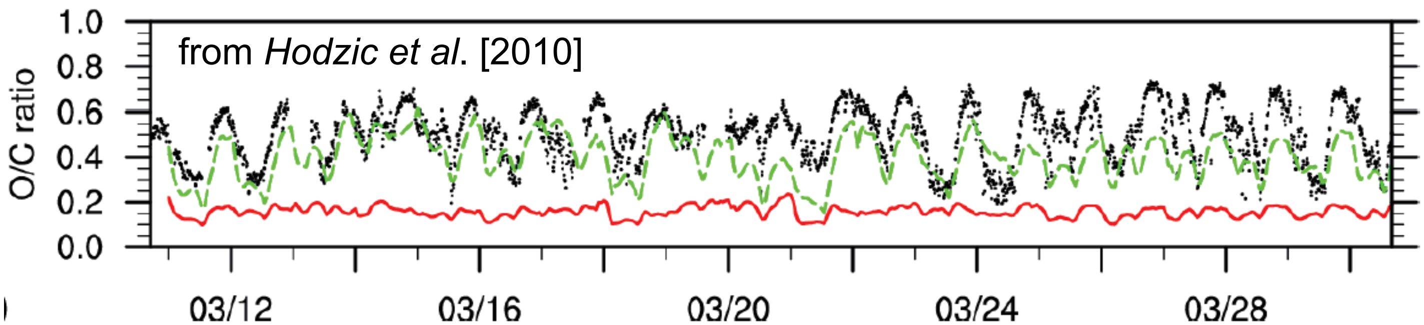
O:C Ratio at T0 Site



Capturing diurnal variation in O/C ratio (higher in afternoon), but still too low

Adding 2 oxygen atoms per generation of oxidation instead of 1



Robinson approach

Data and PMF analysis from Jose Jimenez and Allison Aiken (Univ. CO)

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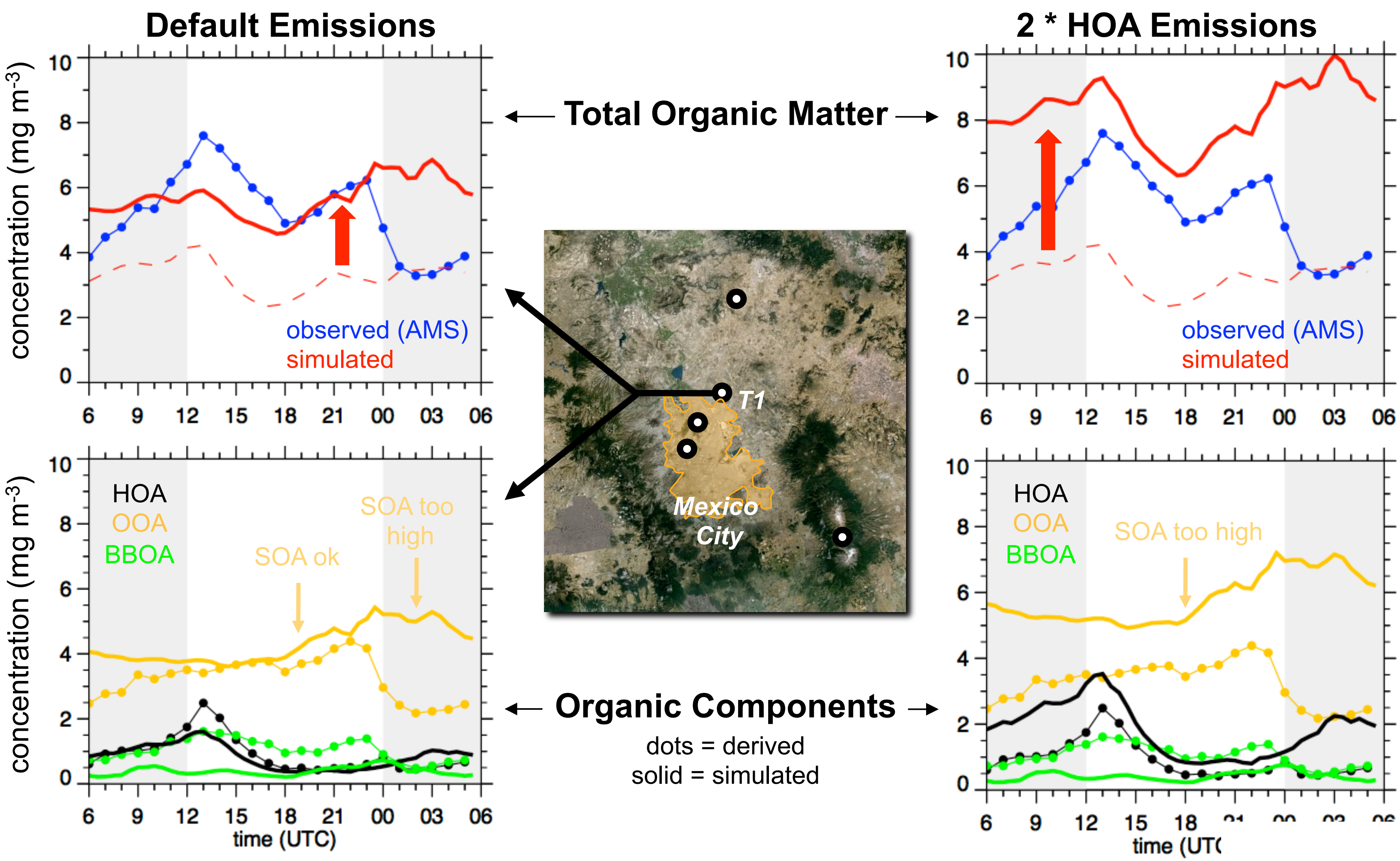
Diurnal Average OM: T1 Site



Data from Liz Alexander (PNNL) and PMF analysis from Manjula Canagaratna (Aerodyne)

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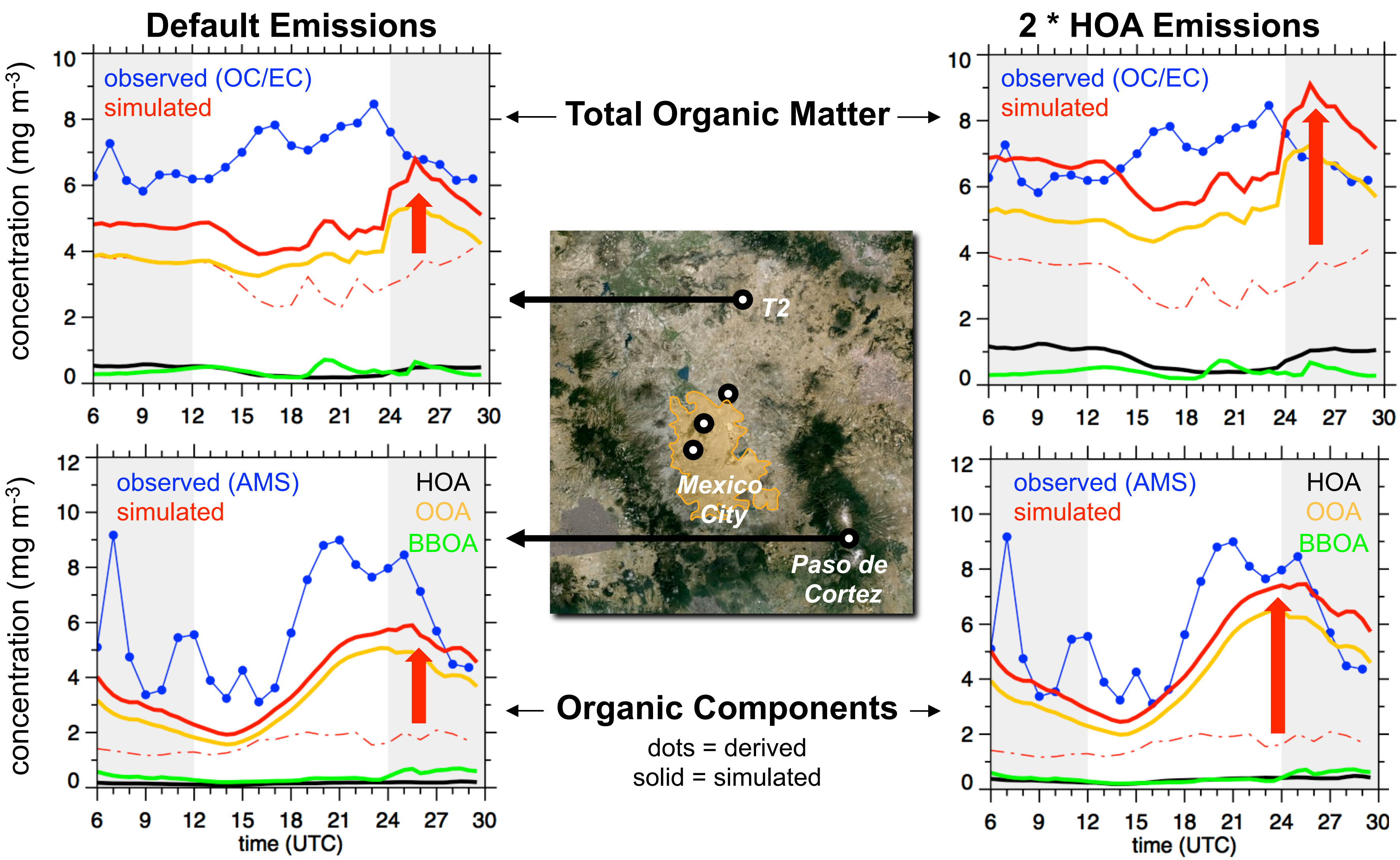
Diurnal Average OM: Remote Sites



Data from Chris Doran (PNNL) and Darrel Baumgardner (UNAM)

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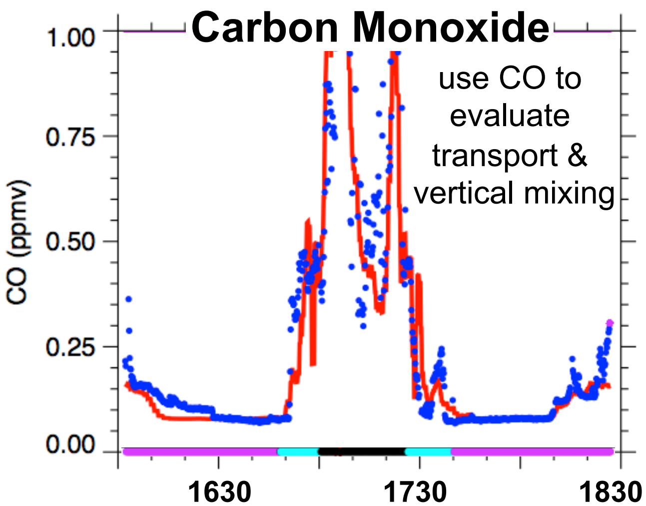
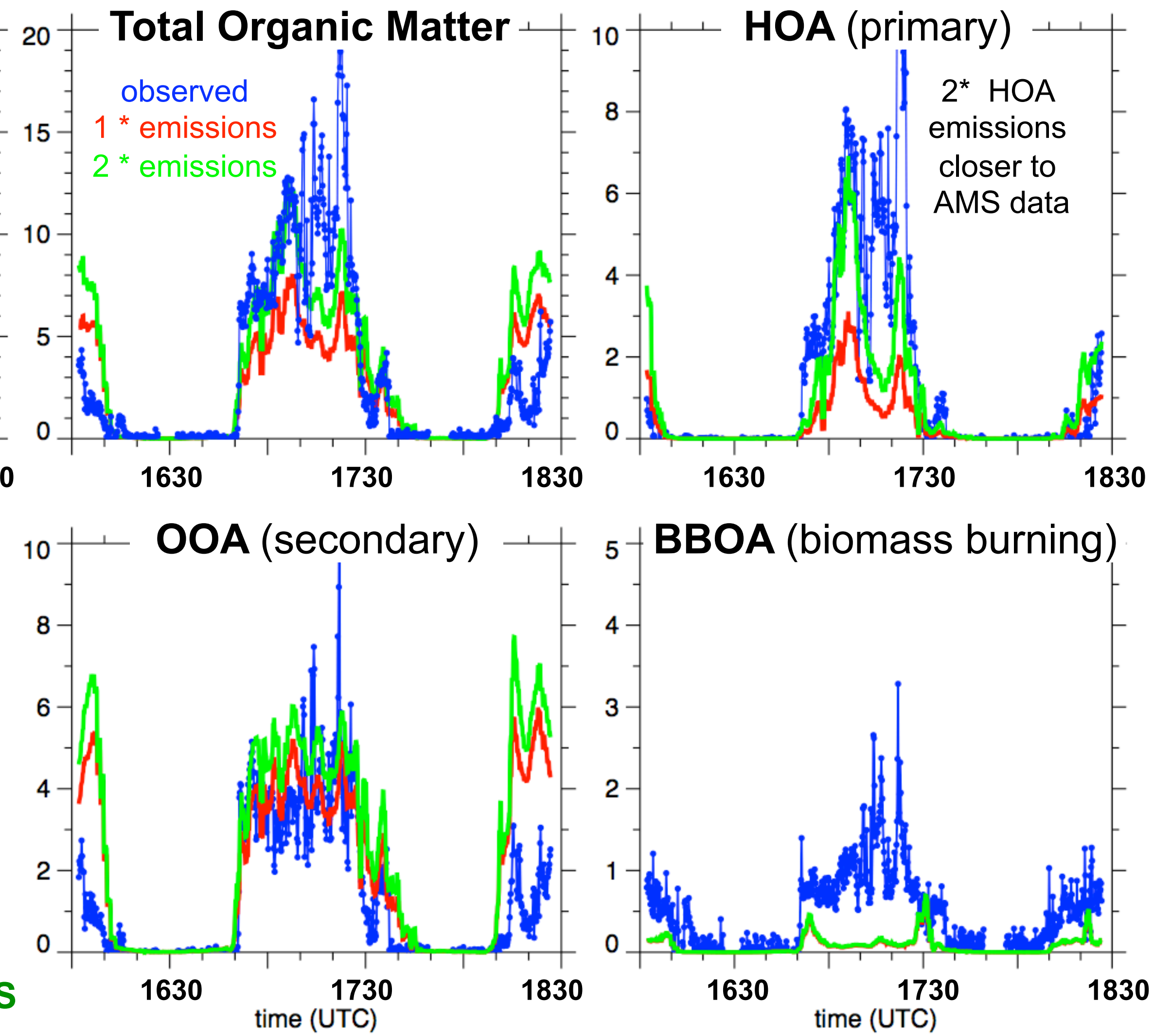
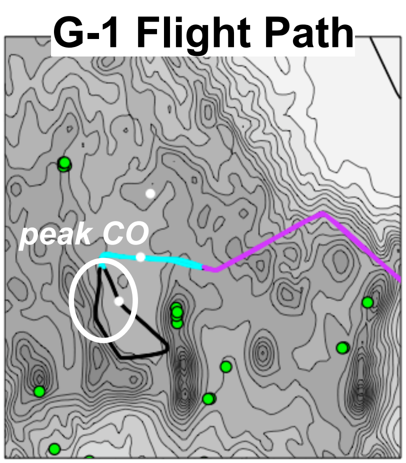
Organic Matter Aloft: March 15







dots = fires detected by MODIS
(low biomass burning day)

AMS data from Liz Alexander (PNNL) and Manjula Canagaratna (Aerodyne)

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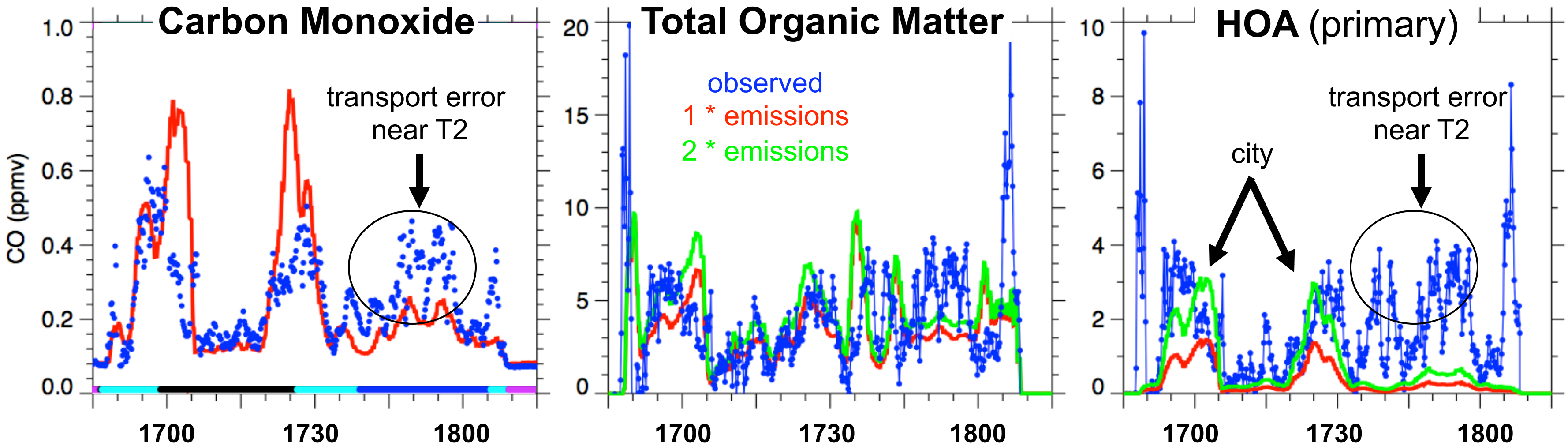
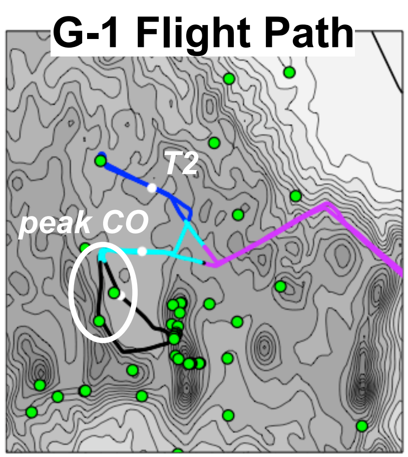
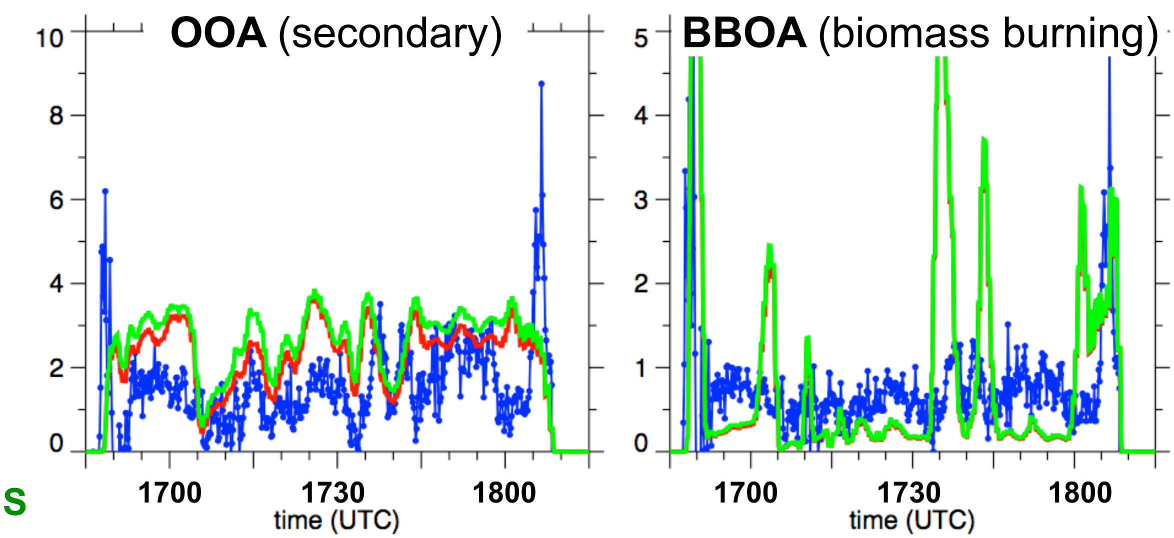
Organic Matter Aloft: March 19







dots = fires detected by MODIS
(high biomass burning day)

AMS data from Liz Alexander (PNNL) and Manjula Canagaratna (Aerodyne)

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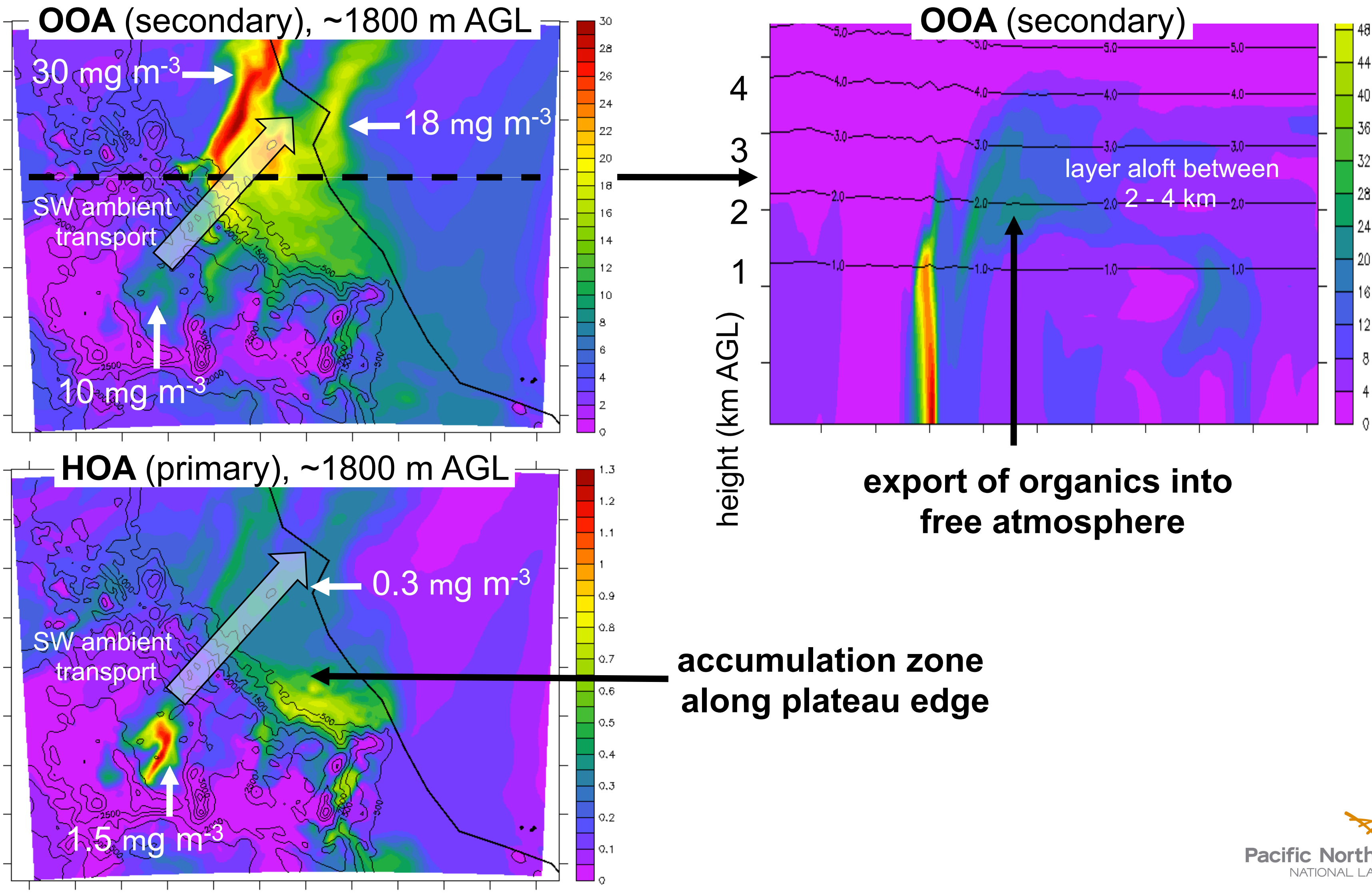
Further Downwind

21 UTC March 10, 2006



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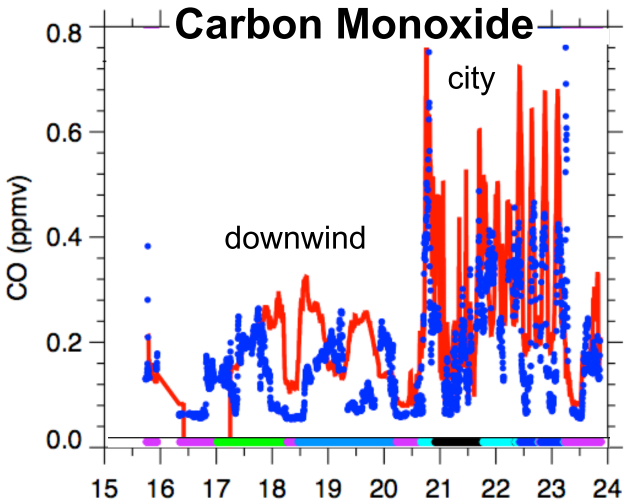
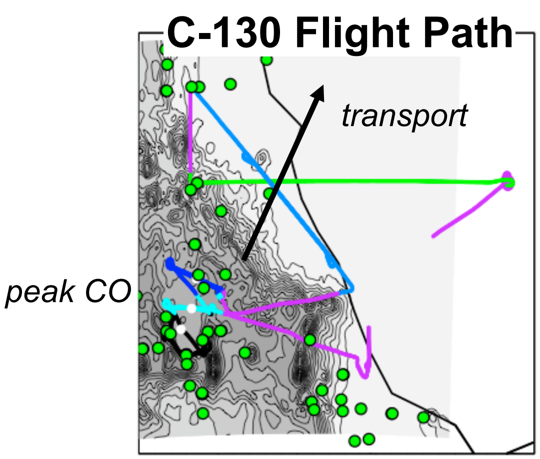
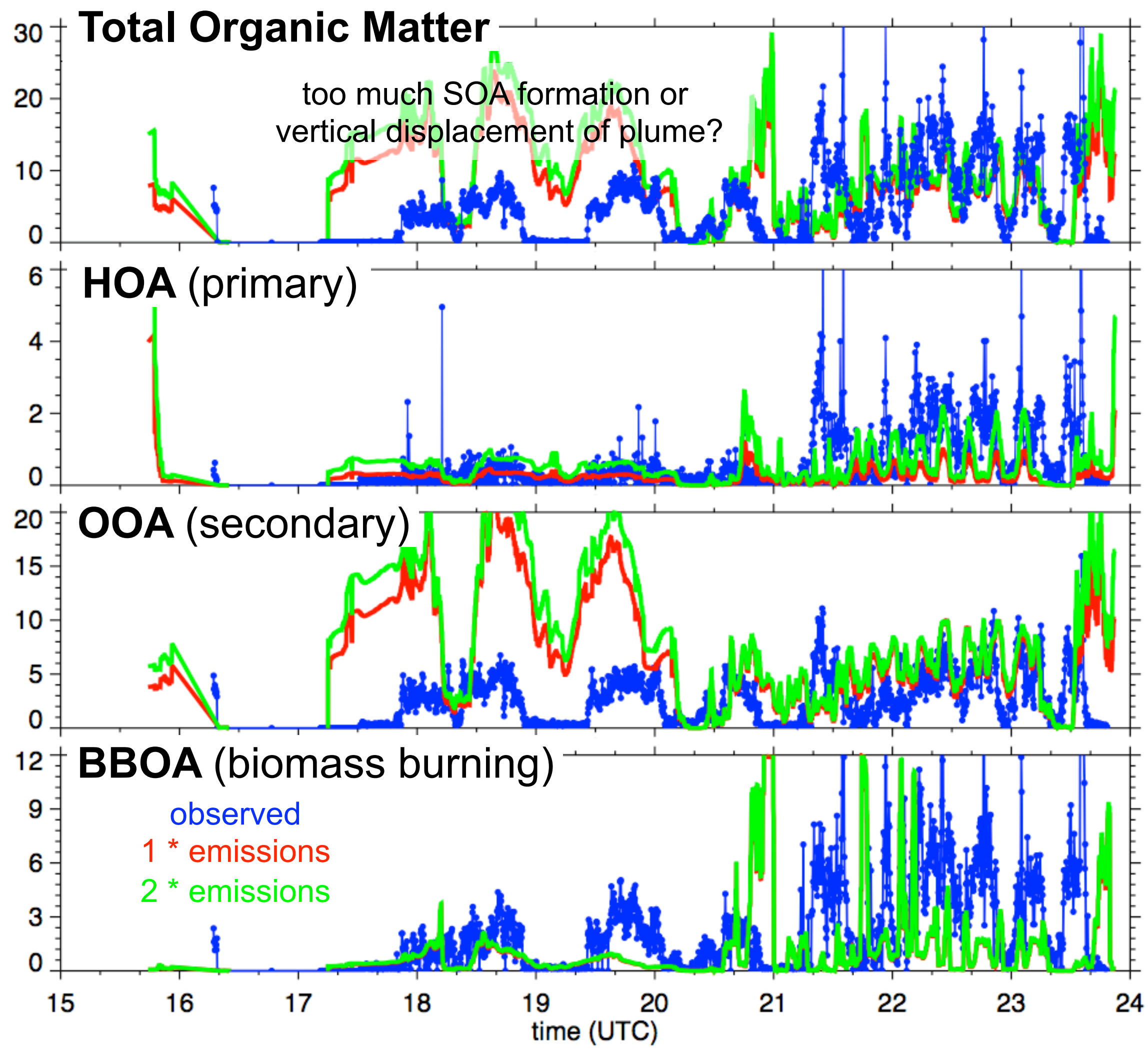
Organic Matter Aloft: March 10





dots = fires detected by MODIS
(high biomass burning day)



AMS data from Pete DeCarlo (PSI) and Jose Jimenez (UC)

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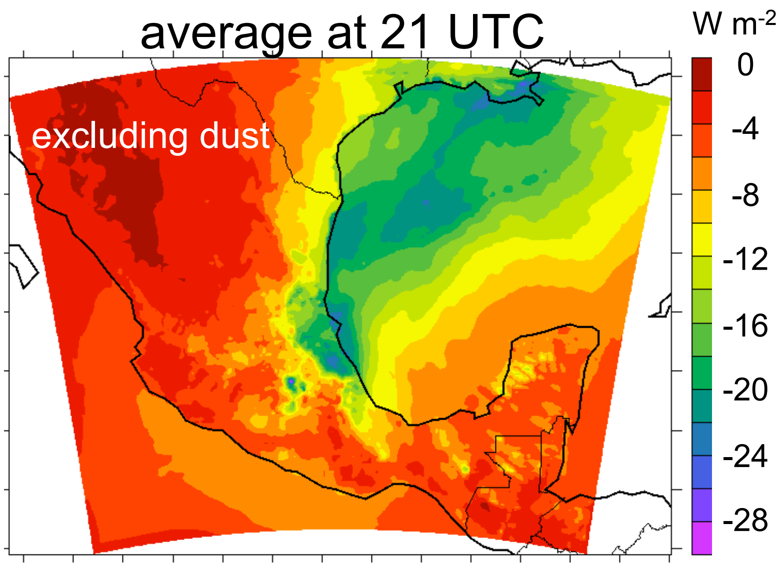
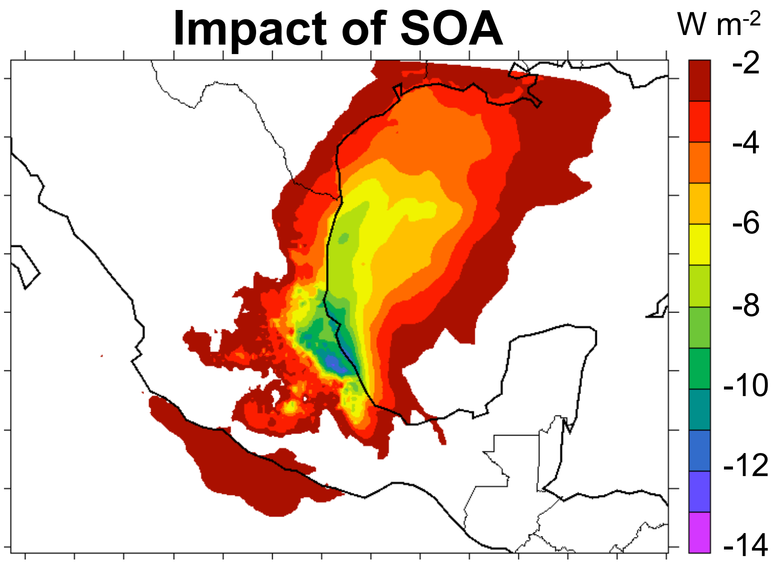
Aerosol Effect on Shortwave Radiation

SW – SW without aerosols
average at 21 UTC

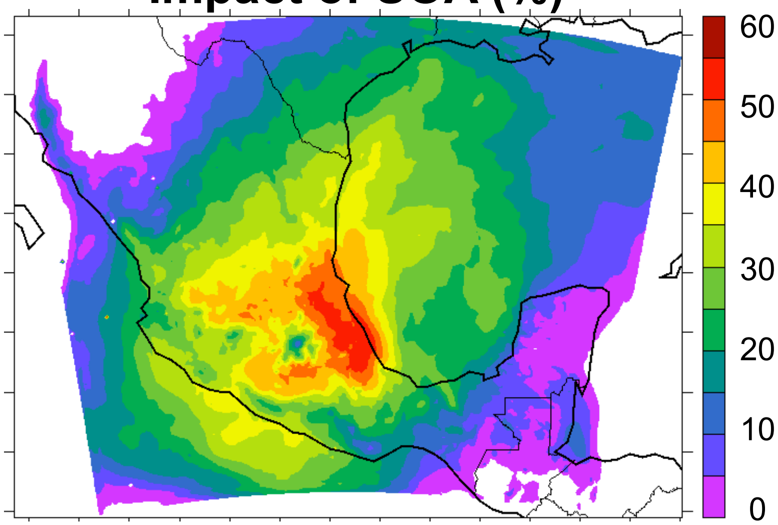


- Aerosols reduce downward shortwave radiation over Mexico City and downwind over the Gulf of Mexico
- SOA contributes to ~50% of reduction in shortwave radiation
- Caveat: simulated OM often too low over the city and too high downwind

Impact of SOA



Impact of SOA (%)



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Summary and Next Steps

- Simulated OM in better than assuming non-volatile POA
- SOA contributed to a large fraction of total reduction in downward SW radiation
- Nevertheless, uncertainties remain:
  - **Primary**: emissions in 2006 inventory likely too low, also uncertainty in spatial variability in emission rates
  - **Secondary**: too low in city, ~observed at downwind site and over the city, but better aloft over city
  - **Biomass Burning**: likely missing sources

} simulated total OM often ~ observed, but compensating errors in components

Next Steps:

- Additional debugging and testing of volatility basis set
- Develop simplified volatility basis set (fewer bins) to ⬇ CPU time (by factor of 3)
- Evaluate using CARES 2010 field campaign data, http://www.arm.gov/campaigns/5793
- Coupling new organic aerosol species with cloud-aerosol interactions

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Acknowledgements:

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- We thank numerous scientists who provided MILAGRO data